Development of Laser Based Techniques for In Situ Characterization of the First Wall in ITER and Future Fusion Devices

V. Philipps[1], A. Malaquias[2], A. Hakola[3], G. Maddaluno[4], P. Gasior[5], M. Laan[6], H.J. van der Meiden[7], M. Rubel[8], A. Huber[1], M.Zlobinski[1], B.Schweer[1], N. Gierse[1], Q. Xiao[1], S. Almaviva[9], L.Caneve[9] , Colao[9], A.Czarnecka[5], M. Kubkowska[5], E.Fortuna[5], P. Petersson[8],

[1]Institute for Energy and Climate Research - Plasma Physics, Forschungszentrum Jülich GmbH, Associat. EURATOM-FZJ, Germany, [2]EFDA-CSU, Culham Science Centre, Abingdon, OX14 3DB, UK and Associação EURATOM/IST, Instituto de Plasmas e Fusão Nuclear, Instituto Superior Técnico, Universidade Técnica de Lisboa, Av. Rovisco Pais 1, 1049-001, Lisboa, Portugal, [3]VTT, Association Euratom-Tekes, P. O. Box 1000, 02044 VTT, Finland, [4]Associazione EURATOM-ENEA sulla Fusione, C. R. Frascati, P.O.Box 65, 00044 Frascati, Roma, Italy, [5]Institute of Plasma Physics and Laser Microfusion, Association EURATOM/IPPLM, 00-908 Warsaw, P.O. Box 49, Hery St. 23, Poland, [6]Institute of Physics, University of Tartu, Association Euratom-Tekes, Tahe 4, Tartu 51010, Estonia, [7]FOM Institute for Plasma Physics Rijnhuizen, Association EURATOM-FOM, Trilateral Euregio Cluster, P.O. Box 1207, 3430 BE Nieuwegein, The Netherlands,[8]Alfvén Laboratory, Royal Institute of Technology, Association EURATOM-VR, 100 44 Stockholm, Sweden, [9] ENEA. UTAPRAD-DIM, P.O.Box 65, 00044 Frascati, Roma, Italy

E-mail contact of main author: v.philipps@fz-juelich.de

Abstract. *In situ* methods to measure the fuel retention and to characterize the material deposition on the wall are highly important for ITER and future fusion devices. Laser-based methods are the most promising candidates (for non-invasive applications) and their feasibility is assessed in a cooperative undertaking in various European associations under EFDA coordination. The work concentrates on three different laser techniques in which the laser light is guided from outside the biological shield by a mirror system through a window onto special wall areas: i) laser induced desorption spectroscopy (LIDS) in which ms laser pulses thermally desorbs the retained fuel from a wall area of about 1cm^2 and this fuel is spectroscopically detected in the edge of a running plasma, ii) laser induced ablation spectroscopy (LIAS) in which ns laser pulses ablates material from a small wall spot and the ablated material together with the incorporated fuel is detected in a similar way as in LIDS and iii) laser induced breakdown spectroscopy (LIBS) in which ns (or even ps) laser pulses produce in front of the irradiated wall spot a plasma plume which (in proper conditions) emits line radiation being a fingerprint of the chemical composition of the ablated materials. The aims are to compare the pros and cons of the methods and propose an optimized solution for ITER. LIDS and LIAS have been developed in the TEXTOR tokamak to a prototype-like status for application in ITER. LIBS has been investigated in several EU laboratories in dedicated lab experiments with a focus on the particular conditions in ITER, including pilot experiments in the TEXTOR tokamak. To enable a clear and fair quantification of the methods, standard deposits of diamond like carbon (DLC) and mixed W/Al/C (with Al mimicing Be) with thicknesses of 2-3 µm deposited on rough and polished W substrates with a known D inventory were prepared using magnetron sputtering and vacuum arc deposition. They were used as reference samples in studies

reported herewith.

1. Introduction

Issues related to plasma-wall interaction processes become increasingly important with the large increase of wall particle fluxes and fluencies and the use of tritium in ITER and in future reactors. This is related at first with the control of the in vessel retention of tritium and more generally with the erosion and subsequent deposition of wall material which mainly determines the T retention via codeposition and at the end the lifetime of first wall components.

Monitoring the wall erosion, material deposition and the associated T- retention is thus of high importance to operate a fusion reactor and in particular to fulfill safety requirements for T retention. Under coordination by EFDA, a special R&D programme is underway in the EU to evaluate the perspectives of different laser techniques to monitor in situ material deposition and T retention for ITER and future reactors. The main results are presented here.

2. Basic description of laser techniques for wall characterization

The basic idea of laser-based diagnostics for in situ characterization of first-wall surfaces in fusion devices is to heat and/or to evaporate material at different spots on the first wall by intensive laser radiation either during or between plasma discharges. Depending on power density and pulse duration, retained hydrogen is released by desorption or wall material and deposits are ablated together with the incorporated hydrogen. Thermally released hydrogen is detected in the edge plasma via neutral hydrogen line emission (Laser Induced Desorption Spectroscopy, **LIDS,**[1,2]). Under ablation conditions, the intense laser radiation produces at first a local laser plasma with an extension of ~1cm and a duration <1µsec, which emits characteristic line radiation. This light is used in the Laser Induced Breakdown Spectroscopy, **LIBS**[3,4,5] for layer analysis. The species leaves the local LIBS plasma and penetrate then into the plasma edge where they emit characteristic line radiation (Laser Induced Ablation Spectroscopy, **LIAS,**[6]). This light has an extension of typically >10 cm in toroidal and poloidal direction and longer lifetime (~ 50 µsec). The line radiation must be measured absolutely and converted into a fluence of released particles based on emission data. The application of LIDS and LIAS require the existence of the main plasma and would ideally be performed in low power plasmas under standard conditions (e.g. in L mode phases before external power application) while LIBS can be used preferentially in between discharges. All three methods have their own advantages and limitations and are thus proposed all together for in situ wall characterization to be used complementary to receive a full picture of the wall status in ITER.

2. Preparation and qualification of "ITER like" deposits.

In order to compare quantitatively the capabilities and limitations of the various laser techniques, ITER-like and other deposits have been produced ex situ on tungsten substrates and their composition and fuel (D) content has been measured ex situ by various techniques, such as RBS , NRA, TDS and SIMS. One class of deposits consisted of carbon films (a-C:H(D) layers) deposited by chemical plasma and arc plasma deposition on top of W with thicknesses ranging from about 0.5 to 3 µm and doped with different amount of D and H. The other deposits were mixtures of W/Al/C (D)(thickness 3µm) -called ITER-like , produced by arc plasma deposition (DIARC) and magnetron sputtering. Al was chosen to mimic Be and the D content ranged from D/C ratios of 0.5 (a-C:D) to a D concentration of 2-4 % (mixed metallic layers). The atomic composition of the mixed layers was determined to be about 50% C, 35% Al and 15% W.

3. Experimental set up for LIBS, LIAS and LIDS in TEXTOR

A test limiter or a sample holder unit sample is introduced via the TEXTOR material test station from the bottom of TEXTOR and positioned at various positions between the LCFS and the wall. On the limiters deposits grow when positioned at distances larger than 1.5 cm outside the LCFS and the ex situ prepared deposits described above were attached to the sample holder. For LIDS, the spots on the target are heated by a laser pulse of 1-3 ms (Nd:YAG laser pulse at 1064 nm) with an area of ~ 0.05-0.1 cm^2 at typically 50-300 kW/cm^2 , or for LIAS and LIBS by an ablation pulse of 7ns duration with an area at the target side of 0.1-0.2 cm^2 and a power of typically 1GW/cm^2. For the LIDS laser, the light is guided by fibre optics (fibre core diameter 400 µm) to a viewport from top of TEXTOR whereby the multiple reflections inside the fibre produce a nearly constant spatial intensity. For ablation, the laser beam (Nd:YAG at 1064 nm) is expanded by a telescope and guided through six dichroic mirrors over a distance of about 35 m into TEXTOR though the same top window, with an optical transmission of the system of ≈ 85% . In order to move the laser spot and the optical observation simultaneously along the surfaces of the test objects, which is also one design proposal for applications in ITER, a prototype of a coaxial laser injection and optical observation system has been built. The laser beam path and the detection path are coaxial and always move together, as shown schematically in Fig 1.

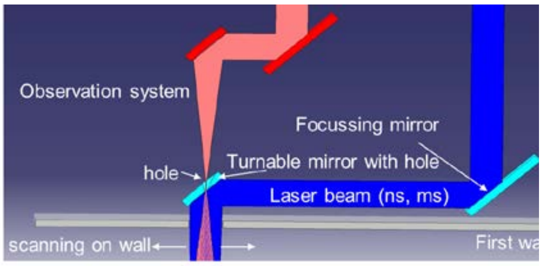

Fig1: schematic view of the coaxial injection and observation system in TEXTOR

The system is built without glass components and replaces lenses by curved metallic mirrors, since glass becomes not transparent by the neutron impact in ITER. The system consists of a curved mirror with an off-axis parabolic shape to allow a beam deflection of 90°to guide the expanded beam of 70 mm and a flat mirror with a diameter of 100 mm both and made of copper with an Al layer and a quartz protection.. To observe LIDS, LIAS and LIBS through the same port, the light passes a 6 mm diameter hole drilled into the second mirror angled at 45°. The light is focused by a mirror of biconical shape onto fibre optics with a 600 µm core diameter and transferred to a spectrometer. To scan the laser spot over the target, the injection and observation system is tilted as a unit by a piezo-driven motor, which allows a precision down to ~0.5 mm in toroidal direction.

With the coaxial observation an area with a diameter of about 23cm around the always centrally located laser spot is recorded by a high resolution spectrometer. In parallel the laser plume is observed via a camera from the top and from the side, to measure the radial and toroidal/poloidal extension of the light emitted. The spectrometer and the cameras are synchronized with the laser pulses, for which typically 4 were applied on the same spot in one TEXTOR shot.

4. LIDS , LIAS and LIBS results

4.1 Laser-Induced Desorption Spectroscopy (LIDS)

Laser-Induced Desorption Spectroscopy has been investigated intensively in TEXTOR in recent years (1,2). It is now validated technique for quantitative analysis of the fuel content. It has been analysed intensively for carbon dominated deposits, also for the mixed deposits used

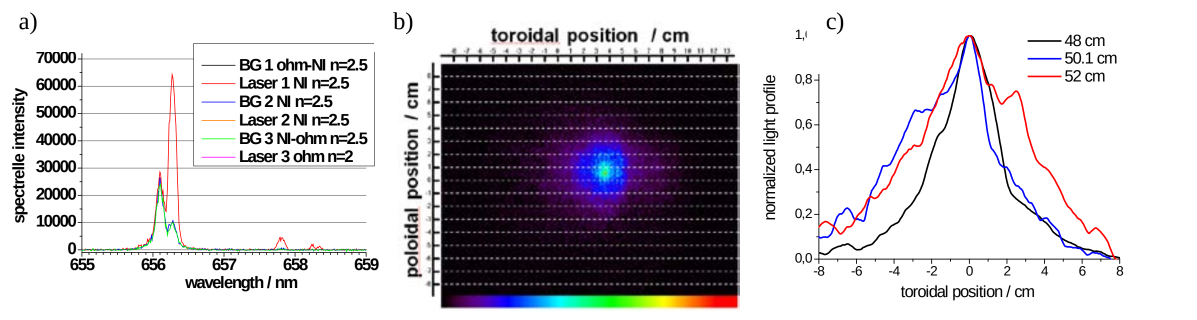

Fig. 2: a) Integrated H, D_α spectrum from the coaxial observation (area ~ 400cm^2, 10 ms) during a LIDS laser shot together with background plasma signal for a 2.5 µm a-C:H deposit on W. b) Top view of the H_α light pattern during the laser shot (background image subtracted). c) Toroidal light distribution for different radial positions of the deposit. The LCFS is at 46 cm.

in this study. A laser pulse of typically 1-3 ms heats the deposit to reach temperatures of typically 1500-2000 K and the released hydrogen is quantified, through the increase of the Hα emission during the duration of the pulse, with respect to the background light in the plasma edge . The light is converted into released H(D) fluxes using absolutely measured photons and conversion factors valid for edge conditions in TEXTOR taken from ADAS database. The laser parameters must be chosen to stay below sublimation/melting ,but reaching temperatures to desorb the vast majority of the retained hydrogen isotopes, if integrity of the wall shall be conserved. For the target positioned 3 cm behind the LCFS, a conversion of 15x2.4 (S/XB x Y to account for molecule desorption) has been used. Fig 2 exemplifies the H, D_α spectrum before and during the laser pulse (10 ms integration) and the Hα pattern from the top view for a LIDS measurement of a 2.5 µm a-C:H deposit on W.

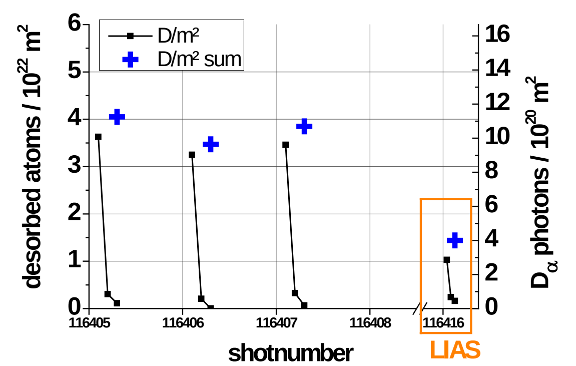

Fig. 3: Collected D_α photons and released deuterium in 3 consecutive TEXTOR shots each with 3 LIDS laser shots on the same deposit (a-C:D layer on C). Ex situ TDS analysis results in 3.8 10^{22} D/m^2. The LIAS data point is discussed below .

Fig 3 shows the spectroscopic signal and the deuterium release in the first 3 laser shots and the cumulative amount (converted into absolute retention using S/XB) for an a-C:D deposit on C in a series of TEXTOR shots. The retained amount of D measured ex situ by TDS was 3.8x10^{22} D/m^2. The shot-to-shot variation on the same deposit (and thus hydrogen release) is about 15% and the agreement of LIDS-analysed fuel content with ex-situ analysis is satisfactory. When carbon dominated deposits are desorbed, a part of the hydrogen is also released in the form of hydrocarbons, which are released at first, followed by hydrogen molecules, which represent the majority (>80%) of desorbed particles.

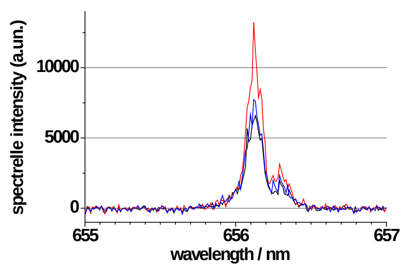

Fig. 4: LIDS and plasma background spectrum of the mixed W/AL/C(D) layers..

The lower detection limit for the method depends mainly on the ratio of the LIDS to plasma background Hα light, which itself depends on the measurement location (high or low recycling area) and the fluctuations of the plasma Hα light. In TEXTOR application from the side view it is 1-5 10^{17} D/cm^2 , but about 10^{18} D/cm^2 for the top view integrating the light over 400 cm^2 for 10 ms, leaving some possibility for improvement by shorter integration in space and time. The release of D from the mixed W/Al/C(D) deposit shows more D released also in the second laser shot, compared with the a-C:D deposits, indicating stronger H bonding. The deuterium content of these layers is low and not much above the lower detection limit, as exemplified in Fig. 4 showing spectra recorded on the spectrum during and before the laser pulse. The deuterium content of the mixed W/Al/C layers has been determined to about 6x10^{21} D/m^2.

4.2 Laser-Induced Ablation spectroscopy (LIAS)

LIAS has been performed in TEXTOR both with a Q switched Ruby laser and later a Nd:YAG laser pulse at 1064 nm with bulk graphite samples (the majority of data), a-C:H(D) layers deposited on C and W (0.5-3 µm) and mixed W-AL-C (D) layers described above. Laser pulses with 7ns, spot areas of 0.1-0.2 cm2 and power densities of 0.5-1 GW/cm^2 have been used. Under such set of experimental conditions, melting and/or sublimation is reached very fast (several tens of a ps) followed by intense material ablation and interaction of the laser with ablated material forming a local laser plasma. This can also be used for characterisation of the ablation (called LIBS) and discussed next. The threshold energy for ablation depends on the material properties and was found to be ~1 J/cm^2 (7 ns) on graphite , ~0.4J/cm^2 for a-C:D deposit on graphite and ~0.25J/cm^2 for a C-deposit (140 nm thickness) on tungsten subtract. With the end of the laser pulse (7ns) the LIBS plasma starts to recombine forming a jet of mainly neutral particles with energies between1eV and 10eV, depending on the laser power density, which penetrate into the edge plasma where they interact with it and emit the LIAS light. This signal is used to characterize and quantify the ablated surface. During the laser shots, the LIBS and LIAS light exists in parallel for some short time but separated in radial direction towards the plasma, as shown e.g. in Fig 5 . The LIBS light extends to about 1cm in front of the laser spot with a volume of about 3-5 cm^3, while the LIAS light is centered in radial direction around the LCFS (46cm in TEXTOR) at ≈ 49-44cm with a toroidal and poloidal extension of about 10cm and a corresponding volume of ≈ 300-500cm^3. This is about 100 times larger than that of the LIBS plasma. The lifetime of LIAS light is about 50 µs, while the LIBS light exist for < 1 µs (under vacuum conditions). Thus LIBS and LIAS can be separated both spatially, but only from suitable viewing directions (e.g. a side view) and temporarily. Fig 6 shows LIAS light for a series of laser shots on an a-C:D deposit on a W target, produced by plasma deposition. The behavior of the CII line shows the removal of the layer in 4-5 shots. Fig 6b shows that the hydrogen release mainly in the first laser shot, but with some remaining hydrogen which is released in subsequent laser ablation shots. Since the heat propagation of a 7 ns heat pulse is about 1µm (in graphite) the laser shots release hydrogen not only by material ablation but simultaneously by thermal release due to the propagating heat wave inside the material. The present analysis shows that the accumulated H_α light (until the interlayer is reached) in LIAS shots which is converted absolutely into hydrogen release using the conversion factors of for TEXTOR (see

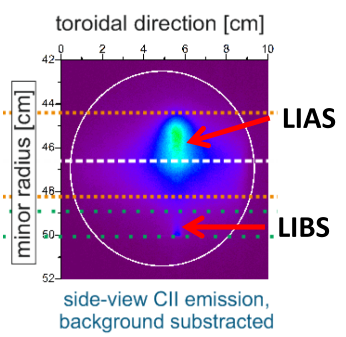

Fig. 5: CII light emission from a tangential view showing LIBS and LIAS light. The deposit is at 50 cm the LCFS at 46 cm..

above) results in less hydrogen when compared with ex situ analysis or also LIDS in situ analysis in TEXTOR (factor of 2-3). This behavior is not fully explained and presently under investigation. It might be due to local change of plasma parameters due to the large particle fluxes of ablated material and/or different release mechanism during desorption and ablation.

4.2 Laser-Induced Breakdown Spectroscopy (LIBS).

The principle of LIBS is explained above. In the frame of the undertaking described here, the major part of the LIBS studies have been performed in laboratory experiments in different EU labs but on the same references deposits (see section 3). Some data exists also for in situ application in TEXTOR and FTU. In these comparative experiments the laser energy densities were matched while the observations differed with respect to wavelength range, detection time window etc. However, the experiments show a good agreement in the ablation rates and the spectroscopic signal intensity ratios. Fig 7(a) shows an example of the shot to shot behavior of the W, C and Al normalized LIBS signals for the mixed deposit, showing that the layer is removed after about 14 laser shots. Fig 7b shows the Dα intensity, which again is characterized by a strong a strong decay after the first laser shot followed by some small remaining signal until the layer is fully ablated after ~14 shots. For LIBS, the most critical part is the conversion of the LIBS signals into absolute numbers of the layer composition and hydrogen content. In most applications this is based on simple comparison of unknown layers with the reference ones. This is also the main strategy for fusion application of LIBS and depends on the amount of empirical knowledge that will be gained on LIBS of “typical” deposits, requiring thus continuous R&D of typical fusion deposits in ex-situ laboratory experiments. However, additional more sophisticated calibration methods are under investigation. One attempt was to compare the photon emission of the unknown deposits with those of the reference bulk materials (C,W,Al, also called slope method, (4)). This gives reasonable results for the fraction of C and W in the mixed deposits from this study ([7]). In addition, absolute photon efficiencies of bulk materials (ratio of emitted photons/ number of ablated atoms) have been evaluated from the absolute photon counting and, the material release from the weight loss and crater depth analysis. Small efficiencies have been found (6): 1-10 x 10^5atoms/photon. The application of these numbers to

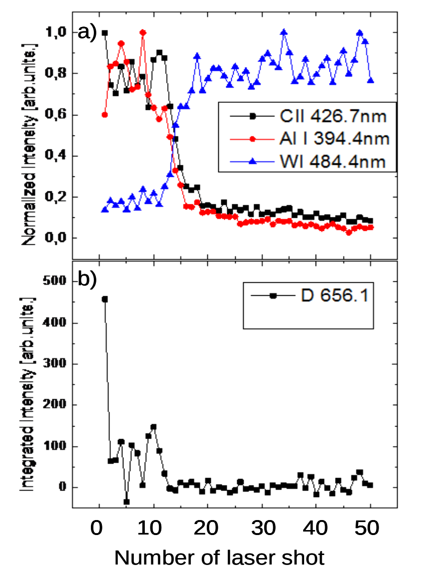

Fig 7: evolution of the W, Al and C line intensities during LIBS from mixed W/Al/C(D) deposit on W (16 J/cm2, 7ns) together with the Dα signal evolution.

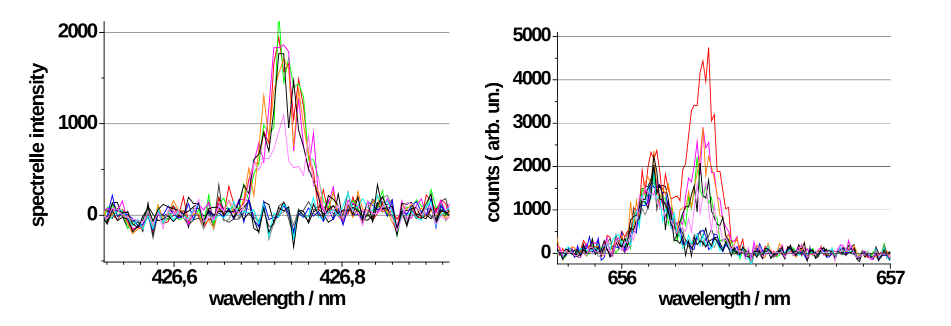

Fig. 6: Integrated (a) CII and (b) D_α signal in TEXTOR during 6 consecutive LIAS laser ablation shots from a-C:H layer deposited on bulk W.

the unknown deposits give some agreement with the real layer composition when ionic emission lines are used, but fails for neutral lines and it is not improved over the simple slope method. So called calibration free methods are also under investigation. They are based on measurements of the temperature and density of the plasma plume derived from Stark broadening (density) and Boltzmann plots of different emission lines (temperature). The density data should indicate whether local equilibrium conditions are achieved (above ~1 x 10^{21} m^{-3}) and the temperature is then used to compare the relative absolute photon intensities with an atomic database which then gives the relative fractions of the materials in the deposits. This method needs accurate atomic data and suffers also from the fast changing plasma parameters during the LIBS plasma lifetime. As such, all the LIBS calibration methods have their pros, cons and limitations. An optimized approach should try to combine empirical calibration with calibration free analysis of emission lines.

LIBS will be analyzed in the near future in more detail in TEXTOR, in particular in comparison with LIAS light emission during the same laser shot on the same deposit. This aims also to investigate the possibility to calibrate in situ the LIBS light, obtained in between shots, with LIAS light generated during plasma operation. Some preliminary analysis has been done on the influence of the magnetic field in TEXTOR and also in FTU on the LIBS signals. The first data show in general some enhancement of the signal with magnetic field, e.g about 40% for a CII line (514nm), but recent measurements indicate a dependence of this enhancement factor on the selected emission line.

5. Implementation in ITER and future devices

The possible application of LIDS, LIAS and LIBS on ITER is affected by many technical constraints. In this study the conditions of a port plug laser system has been explored, while a possible system mounted on a remote arm (which can then only be LIBS of course) is outside this study. The reasons to concentrate on a port plug system are connected with the larger complexity of a remote arm LIBS system, its restriction for application to remote handling operation and the advantage of a port plug system to combine LIDS, LIAS and LIBS. For such a system based on a port plug, the laser beam should heat a wall area of about 1 cm^2 and the emitted light should be observed in a coaxial way both emitted in edge plasma (LIDS and LIAS) and from LIBS, preferably in between plasma operation. Detection of LIAS and LIDS light by other existing spectroscopic diagnostics can support the analysis. A movable mirror should provide laser scanning of the inner wall, the upper dump plates and a section of the upper part of the inner divertor and dome. This is one of the major technical challenges of this concept. A preliminary system for a coaxial observation and movable mirror and detection has been built and operated in TEXTOR. Based on this an upgrade has been designed which reflects more the needs for application in ITER. The moving parts of the last mirror must operate under vacuum and the laser should allow impact angles down to ≈30° and also some variations in the focus of the laser due to varying distances to the wall. On the ITER port, the opening for laser beam steering and collection of the spectral light should be < 100 cm and based fully on a mirror system, while the application of fibers and other refractive optical components is excluded due to neutron and plasma impact. Therefore, low divergence lasers (<0.5mrad) are needed to transport the light by reflective optics (mirror system) over long distances (>50m) to the final focusing mirror in ITER.

Several B2-Eirene modeling has been done to model the LIAS and LIDS signals expected in ITER. From the spot of 1 cm^2 a hydrogen flux of 3×10^{21}H/m^2 was released in 1ms with a Maxwellian energy distribution into the edge plasma. The spatial extension of Hα light intensity of the desorbed hydrogen was calculated with B2-EIRENE and compared with the intrinsic hydrogen light. The analysis shows that the signal to background ratio of such a source at the inner wall is about ten, providing enough margins to detect it, but decreases when moving down the inner wall to the upper baffle divertor region, due to increasing background Hα light. At the lower end of the upper inner baffle, the detection limit is estimated to be 10^{22} D/m^2. For LIBS the number of photons has to be considered. With an estimated solid angle of $\approx 2\times10^{-6}$sr, a transmission factor and detector yield of 0.1 and an effective photon efficiency of 10^5-10^6 released atoms/photon, as measure din lab experiments, about 10^{18}C atoms) must be ablated to obtain enough statistics.

6. Conclusions

R&D is ongoing to qualify laser based methods for the in-situ characterization of the wall composition (material deposition) and fuel retention in ITER and future devices. LIDS is based on thermal release of hydrogen from the laser spot with its spectroscopic detection in the edge plasma. This technique has been qualified in TEXTOR and proven to provide reliable absolute amount of fuel retention from carbon like and mixed deposits. However, it relies on the total desorption of fuel during the laser shots and has its limitations on the detection of hydrogen which is more deeply (> 50 µm) penetrated inside the bulk of wall components.. LIAS is based on ablation in the laser spot area and detects also both hydrogen and ablated material spectroscopically in the edge plasma. Its absolute quantification needs additional analysis, in particular to assess the effect of the ablated material on the local plasma which affects the conversion of the observed light into the absolute amount of released particles.

LIBS should be applied between plasma shots (thus with magnetic field on and under high vacuum). It is well suited to detect qualitatively the composition of deposits and to identify the interlayer to the bulk material and, by this, the interlayer thickness. The ablation rates used in the assessment can be defined with good accuracy in ex situ laboratory experiments. The determination of the absolute composition of the deposit and its hydrogen content with LIBS is more challenging. It must be based on a combination of an empirical approach, for which unknown deposits in fusion are compared with reference layers which are analysed ex situ and “calibration free “ methods which analyse LIBS plasma parameters (ne, Te) and compare emission lines with atomic databases.

To optimize the analysis of in-situ wall composition and fuel retention by laser based techniques it is proposed to combine LIBS with LIDS and LIAS to enable both spectroscopy of emitted species by the LIBS plasma and in the edge of a running fusion plasma.

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